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Life cycle human health impacts of 875 pesticides

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Abstract

Purpose Residues in field crops grown and harvested for human consumption are the main contributor to overall human exposure toward agricultural pesticides for the general population. However, exposure from crop residues is currently not considered in life cycle assessment practice. We therefore present a consistent framework for characterizing human toxicological impacts associated with pesticides applied to agricultural crops in the frame of life cycle impact assessment based on state-of-the-art data and methods.

Methods We combine a dynamic multicrop plant uptake model designed for evaluating human exposure to residues for a wide range of pesticide-crop combinations with latest findings of pesticide dissipation kinetics in crops and post-harvest food processing. Outcome is a set of intake fractions and characterization factors for 875 organic pesticides and 6 major food crops along with specific confidence intervals for each factor.

Results and Discussion Intake fractions aggregating exposure via crop residues and exposure via fractions lost to air and soil for pesticides applied to agricultural crops vary between 10^{-8} and 10^{-1} kg intake per kg applied as a function of pesticide and crop. Intake fractions are typically highest for lettuce and tomato and lowest for potato due to differences in application times before crop harvest and soil as additional barrier for uptake into potato tubers. Uncertainty in intake fractions is mainly associated with dissipation dynamics in crops, where results demonstrate that using pesticide- and crop-specific data is crucial. Combined with the uncertainty in effect modeling, characterization factors per pesticide and crop show squared geometric mean standard deviations ranging from 38 to 15560 over a variability range across pesticide-crop combinations of 10 orders of magnitude.

Conclusions Our framework is operational for use in current life cycle impact assessment models, is made available for USEtox, and closes an important gap in the assessment of human exposure to pesticides. For ready use in life cycle assessment studies, we present pesticide-crop combination-specific characterization factors normalized to pesticide mass applied and provide default data for application times and loss due to post-harvest food processing. When using our data, we emphasize the need to consult current pesticide regulation, since each pesticide is registered for use on certain crops only, which varies between countries.

Keywords: dynamiCROP plant uptake model; human toxicity characterization factors; pesticides; life cycle impact assessment (LCIA); food crop consumption; intake fractions

1 Introduction

Food is an important source of human exposure to toxic chemicals which includes residues of pesticides, perfluorinated chemicals, metals, phthalates, and persistent organic pollutants including dioxins and polychlorinated biphenyls. Common sources of residues in food along food product life cycles are agricultural production and harvesting, food packaging, storage, industrial and domestic food processing, and finally serving (Dickson-Spillmann et al. 2009, Freeman 2011, Lippmann 2009, Muncke 2009, Tittlemier et al. 2007). In this context, pesticides are a special chemical class of interest, because they are intentionally applied to agricultural field crops, they have by design toxic properties, and the general public in various countries is concerned about chronic effects from low-level exposure (European Commission 2006, McKinlay et al. 2008, Pretty 2005, Slovic 2010). For pesticides, food crop consumption is the predominant pathway for human exposure (Caldas & Jardim 2012, Fantke et al. 2011a, Lu et al. 2008). Therefore, assessing pesticide residues in food crops is a key component in current pesticide authorization in Europe (European Commission 2009) and elsewhere and needs to be considered for assessing the environmental performance of food products over their life cycle.

Life cycle assessment (LCA) is a tool that is frequently applied to evaluate the environmental performance of agricultural production systems as well as various food products including crops (Andersson 2000, Perrin et al. 2014, Roy et al. 2009, Schau & Fet 2008). However, although health impacts from environmental emissions associated with the use of pesticides in food crop production are considered in some agrifood-related LCA studies, human exposure to pesticide residues in the treated food crops is still mostly disregarded (Fantke et al. 2011b, Juraske & Sanjuán 2011). This is mainly due to the fact that current tools for estimating pesticide residues in food crops show considerable uncertainties – mostly associated with dissipation kinetics in crops (Fantke et al. 2012a, Juraske et al. 2008). Furthermore, these tools are not implemented in current life cycle impact assessment (LCIA) models and methods for assessing human health impacts from exposure to potentially toxic chemicals including pesticides. To address this gap, we propose to (a) develop an operational framework for consistently incorporating health impacts from exposure to residues in food crops associated with field applications of agricultural pesticides into LCIA. We further aim at reducing uncertainty of pesticide-related characterization factors by integrating the latest findings from Fantke and Juraske (2013) and Fantke et al. (2014) in estimating dissipation kinetics in crops to (b) calculate harvest fractions, intake fractions and characterization factors for 875 pesticides and to (c) estimate the resulting specific uncertainty for each of these factors.

2 Methods

The general framework applied in LCIA for characterizing human toxicological impacts associated with chemical emissions combines factors representing environmental fate, human exposure, and health endpoint-specific dose-response into characterization factors (European Commission 2010, Udo de Haes et al. 2002). At midpoint level, human toxicological characterization factors relate numbers of health incidences to emitted chemical mass. At endpoint level, characterization factors contain an additional term accounting for the (damage or health endpoint-specific) severity and are expressed in terms of disability-adjusted life years (DALY) per emitted chemical mass. Environmental fate and human exposure can be aggregated into the human intake fraction that directly relates the chemical mass taken in by an exposed (or the entire global) human population to the chemical mass emitted (Bennett et al. 2002). This general framework for assessing human toxicity impacts in LCIA under assumed steady-state conditions was originally designed to be applied for environmental emissions, i.e. related characterization factors are normalized to a unit mass continuously released into a specific environmental compartment, such as air, water, or soil (Rosenbaum et al. 2008). However, pesticides are not emitted continuously, but are rather applied as pulses to agricultural crops that are harvested within days to weeks after the (latest) application. Steady-state might, hence, often not be reached, especially when pesticides are applied shortly before crop harvest (Fantke et al. 2013, Rein et al. 2011). In addition, the fraction of the applied pesticide mass that is intercepted by the crop surface and that ends up as residues in crop harvest along with the fractions that are lost during and after the application and that reach target field and off-target soil, air and water including surface and groundwater are not typically reported or available for LCA practitioners (Perrin et al. 2014). Instead, in most cases the applied pesticide mass or mass per area is available, from which fractions reaching the treated crop and fractions reaching the environment as emissions then need to be estimated (Rosenbaum et al. 2015). Consequently, the current framework applied for human toxicity assessment of chemicals in LCIA needs to be extended and modified as detailed in the following to reflect the mass distribution dynamics between pesticide application and food crop harvest.

2.1 Modeling framework for pesticide exposure

Characterization factors: Our starting point is the multicrop model for characterizing health impacts from pesticide residues in food crops, dynamiCROP, that describes the mass evolution of pesticides in different crop-environment systems based on solving a set of coupled differential equations. This model is fully described in Fantke et al. (2011a) and Fantke et al. (2011b) and is designed for evaluating human toxicological impacts associated with pesticide residues in wheat, paddy rice, apple, tomato, potato and lettuce, representing the most relevant crop archetypes with respect to human vegetal food consumption. Following this approach, human toxicity characterization factors, $\mathrm{CF}_{x,t,e}$ [incidences $\mathrm{kg}_{\mathrm{applied}}^{-1}$ at midpoint level, $\mathrm{DALY\,kg}_{\mathrm{applied}}^{-1}$ at endpoint level], for pesticides applied to crop *x* harvested at time *t* [days after application] associated with health endpoints *e* are calculated from toxicity effect factors for aggregated cancer and non-cancer health effects, EF_e [incidences $\mathrm{kg}_{\mathrm{intake}}^{-1}$ at midpoint level, $\mathrm{DALY}\,\mathrm{kg}_{\mathrm{intake}}^{-1}$ at endpoint level] and human intake fractions, $\mathrm{iF}_{x,t}$ [$\mathrm{kg}_{\mathrm{intake}}\,\mathrm{kg}_{\mathrm{applied}}^{-1}$], as

$$\mathrm{CF}_{x,t,e} = \mathrm{EF}_e \times \mathrm{iF}_{x,t} \tag{1}$$

Effect factors: Effect factors are derived as

$$\mathrm{EF}_e = \begin{cases} \mathrm{DRF}_e & \Rightarrow \text{midpoint level} \\ \mathrm{DRF}_e \times \mathrm{SF}_e & \Rightarrow \text{endpoint level} \end{cases} \tag{2}$$

with DRF_e [incidences $\mathrm{kg}_{\mathrm{intake}}^{-1}$] as dose-response slope factor and SF_e [$\mathrm{DALY}\,\mathrm{incidence}^{-1}$] as damage or severity factor. Dose-response slope factors relate risks of humans to potentially develop a health effect from pesticide exposure to the quantity inhaled or ingested and are mainly taken from Rosenbaum et al. (2008). In case of missing data, DRF_e are extrapolated from chronic lifetime doses affecting 50% of exposed humans or – if chronic data are not available as for most non-cancer effects – from no-observed effect levels of exposed animal species assuming linear dose-response relationships (Huijbregts et al. 2005, Kramer et al. 1996). The difference in the units of the effect factors (Eq. 2) and consequently of the characterization factors is related to the fact that at midpoint level, the effect factor is solely derived from (and therefore equal to) the dose-response slope factor, whereas at endpoint level, a severity factor is included accounting for differences in effect severity. Severity factors of 11.5 and 2.7 $\mathrm{DALY}\,\mathrm{incidence}^{-1}$ are applied for cancer and non-cancer effects, respectively (Huijbregts et al. 2005), to be used for comparative purposes rather than for estimating absolute damages. Disability-adjusted life years are undiscounted and without age-weighting.

Human intake fractions: To account for both the pesticide mass fraction reaching the crop as residues and the fractions lost as emissions to air and soil during and after application, human intake fractions relate the mass that is ultimately taken in by humans via all exposure pathways to the mass of applied pesticide. Hence, the total intake fraction per mass applied combines the specific intake fractions for exposure to crop residues from the applied mass reaching the treated crop with intake fractions for different exposure pathways *p* including inhalation and ingestion of drinking water and different food items from the applied mass reaching air and soil:

$$\mathrm{iF}_{x,t} = \mathrm{iF}_{x,t}^{\mathrm{residues}} + fr_x^{\mathrm{air}} \times \sum\nolimits_p \mathrm{iF}_p^{\mathrm{air}} + fr_x^{\mathrm{soil}} \times \sum\nolimits_p \mathrm{iF}_p^{\mathrm{soil}} \quad (3)$$

where $\mathrm{iF}_{x,t}^{\mathrm{residues}}$ $[\mathrm{kg_{intake}\ kg_{applied}^{-1}}]$ is the intake fraction associated with exposure to residues in the treated crop at harvest time based on a mechanistic plant uptake model accounting for partitioning, transport and dissipation kinetics (see Section 2.2), $\mathrm{iF}_p^{\mathrm{air}}$ $[\mathrm{kg_{intake}\ kg_{emitted\ to\ air}^{-1}}]$ is the exposure pathway-specific intake fraction related to the fraction lost to air via e.g. wind drift, fr_x^{air} $[\mathrm{kg_{emitted\ to\ air}\ kg_{applied}^{-1}}]$, and $\mathrm{iF}_p^{\mathrm{soil}}$ $[\mathrm{kg_{intake}\ kg_{emitted\ to\ soil}^{-1}}]$ is the exposure pathway-specific intake fraction related to the fraction lost to soil via e.g. deposition, fr_x^{soil} $[\mathrm{kg_{emitted\ to\ soil}\ kg_{applied}^{-1}}]$. Intake fractions referring to mass emitted to air (beyond treated field boundaries) and to soil (considering run-off and leaching to freshwater and disregarding direct crop uptake as already considered in the intake fractions related to crop residues) are calculated with USEtox (Rosenbaum et al. 2008) assuming continuous emissions and steady-state conditions. Related fractions lost (emitted) to air during and after pesticide application are assumed to be pesticide-generic, but crop-specific for a typical foliar application and range from 5% for lettuce, 14.8% for potato, and 16.5% for wheat and paddy rice to 23.7% for tomato and 35.4% for apple (Ganzelmeier et al. 1995, Rautmann et al. 2001, van de Zande et al. 2007). We thereby acknowledge that different application techniques, such as aerial or soil application would yield different fractions lost to air. Fractions lost to soil range from 2.3 to 81% assuming foliar application and are a function of pesticide properties (e.g. molecular weight influencing deposition velocities), application time (where we distinguish per crop between averages for herbicides and other pesticides), and crop characteristics (e.g. growth stage and interception area influencing intercepted pesticide mass).

2.2 Exposure to residues in crops

Intake fractions for crop residues are calculated from harvest fractions representing the residual mass fraction of applied pesticide found in crop harvest, $\mathrm{hF}_{x,t}$ $[\mathrm{kg}_{\text{in harvest}}\ \mathrm{kg}_{\text{applied}}^{-1}]$, and a food processing factor, PF_x $[\mathrm{kg}_{\text{intake}}\ \mathrm{kg}_{\text{in harvest}}^{-1}]$, accounting for post-harvest reduction of crop residues due to subsequent food processing steps:

$$\mathrm{iF}_{x,t}^{\text{residues}} = \mathrm{PF}_x \times \mathrm{hF}_{x,t} \tag{4}$$

Since data are only available for a limited number of pesticide-crop combinations, pesticide-generic food processing factors are applied as proxies, i.e. 0.59 $\mathrm{kg}_{\text{intake}}\ \mathrm{kg}_{\text{in harvest}}^{-1}$ for washing with tap water, 0.31 $\mathrm{kg}_{\text{intake}}\ \mathrm{kg}_{\text{in harvest}}^{-1}$ for parboiling or cooking, and 0.33 $\mathrm{kg}_{\text{intake}}\ \mathrm{kg}_{\text{in harvest}}^{-1}$ for bread making (Kaushik et al. 2009, Keikotlhaile et al. 2010, Liang et al. 2014).

Harvest fractions are defined as the ratio of residual pesticide mass in all harvested crop components *c*, $m_{x,c,t}^{\text{residues}}$ $[\mathrm{kg}_{\text{in harvested crop component}}]$, and the sum of applied pesticide mass, m_x^{applied} $[\mathrm{kg}_{\text{applied}}]$, and background mass, $m_x^{\text{background}}$ $[\mathrm{kg}_{\text{in crop-environment system}}]$:

$$\mathrm{hF}_{x,t} = \frac{\sum_c m_{x,c,t}^{\text{residues}}}{m_x^{\text{applied}} + m_x^{\text{background}}} \approx \frac{\sum_c m_{x,c,t}^{\text{residues}}}{m_x^{\text{applied}}} \tag{5}$$

Both the pesticide mass applied directly to the treated crop and the soil residues from deposition or from earlier applications (background mass) that are taken up into the crop via the root system need to be considered according to Eq. 5. However, following the FAO recommendations for good agricultural practices for pesticide application (FAO 2003) as best estimate in LCIA, we assume that the background pesticide input via root uptake from previous applications and/or cross-field wind drift and subsequent deposition onto soil are negligible, i.e. we assume $m_x^{\text{background}} \approx 0$ kg. We justify this assumption with the fact that even when applied in relatively quick succession to the same crop, previous studies have demonstrated that typically only the latest direct application is dominating overall residues in crop harvest (Juraske et al. 2011, Rein et al. 2011). Hence, harvest fractions and all subsequent metrics, i.e. intake fractions and characterization factors, are normalized to the (latest) pesticide mass applied to the respective crop, m_x^{applied}.

To obtain harvest fractions, we have solved the dynamics of a mass balance system of environmental compartments including air, soil and paddy water (the latter only for paddy rice) and crop components including root, stem, leaves, leaf surface, fruit and fruit surface (the latter two for all crops but lettuce and potato), which are all coupled by inter-compartment transfers (Fantke et al. 2011a, Rein et al. 2011). Crop residues and resulting harvest fractions were found to be highly dependent on degradation in crops and time to harvest (Fantke et al. 2012b). From comparing modeled crop residues with measured data, we found that predicted residual masses over time were in good agreement with measured residues with R^2 between 0.81 and 0.99 (Fantke et al. 2011a, Fantke et al. 2011b, Itoiz et al. 2012, Juraske et al. 2012). Since most input parameters that are required for solving the underlying mass balance system are typically not available to LCA practitioners and to be compatible with the format of assessment models and intake fractions applied in LCIA for human toxicity assessment, the dynamiCROP model was linearized and a parametric regression model was developed for each of the six crops still accounting for the main influences on the dynamics between pesticide application and crop harvest (Fantke et al. 2012b, Fantke et al. 2013). Each model combines the contributions of different crop and environmental components $c \in \{\text{crop interior, crop surface, soil}\}$ at harvest time to the overall residual pesticide mass found in crop harvest:

$$\mathrm{hF}_{x,t} = \sum_c \mathrm{hF}_{x,c,t} = \sum_c 10^{(\alpha^*_{x,c} + \beta_{x,c} \times k_{x,c} \times t_x)} \qquad (6)$$

where $\alpha^*_{x,c}$ and $\beta_{x,c}$ denote dimensionless coefficients, $k_{x,c}$ $[\mathrm{kg}_{\text{reaching component}}\,\mathrm{day}^{-1}$ per $\mathrm{kg}_{\text{in component}}]$, represent removal rate coefficients and t_x [day] is the time between pesticide application and crop harvest. Crop- and crop/environmental component-specific coefficients $\alpha^*_{x,c}$ and $\beta_{x,c}$ are detailed in the Supporting Information (SI), Section S-1, and are adapted from Fantke et al. (2012b). Removal rate coefficients for the soil component $k_{x,c\in\{\text{soil}\}} = 1/\mathrm{FF}_{\text{soil}}$ are derived from the inverse of pesticide residence times in soil corresponding to the fate factors for continental agricultural soil, $\mathrm{FF}_{\text{soil}}$ $[\mathrm{kg}_{\text{in soil}}$ per $\mathrm{kg}_{\text{emitted to soil}}\,\mathrm{day}^{-1}]$, in the USEtox matrix of fate factors (Rosenbaum et al. 2008). Removal rate coefficients for crop interior and crop surface are generally obtained as $k_{x,c\in\{\text{crop;crop-surface}\}} = \ln(2)\big/\mathrm{HL}^{\text{dissipation}}_{x,c}$ from overall removal (dissipation) half-lives $\mathrm{HL}^{\text{dissipation}}_{x,c}$ [day] estimated by Fantke et al. (2014) by fitting dissipation kinetics for 1485 distinct pesticide-crop combinations reported in Fantke and Juraske (2013). For tomato, apple and lettuce, additional terms contribute to $k_{x,c\in\{\text{crop;crop-surface}\}}$ accounting for the influence of substance properties (see SI, Section S-1). Finally, crop-specific harvest times are taken from Fantke et al. (2011b), Table S1, separately averaged for herbicides typically applied before or during early crop stages and other pesticides, such as fungicides and insecticides, applied during all crop stages including shortly before harvest and during post-harvest storage. With these assumptions, we yield best estimates for crop residues and typically do not exceed regulatory maximum residue limits (MRL) as demonstrated by Juraske et al. (2011), Juraske et al. (2012), Itoiz et al. (2012), and Fantke et al. (2011a).

2.3 Uncertainty analysis

Uncertainty of harvest fractions, intake fractions and characterization factors (model output) is expressed as 95% confidence interval ranges. Confidence intervals around model output y are derived from a combination of uncertainty related to model input variables (input parameter uncertainty) and uncertainty related to modeling of harvest fractions (regression model uncertainty). Input parameter and model uncertainty are expressed as squared geometric standard deviations $\mathrm{GSD}_{x_i}^2 := \exp(2\times\sigma_{x_i})$ with $\sigma_{x_i} > 0$ the standard deviation of the natural logarithm of input variable or regression model x and the probability $\{x_i / \mathrm{GSD}_{x_i}^2 < x_i < \mathrm{GSD}_{x_i}^2 \times x_i\} = 0.95$ representing the 95% confidence interval around x:

$$\mathrm{GSD}_y^2 = \exp\left(2\times\sqrt{\sum_i \mathrm{var}(\ln(x_i))}\right) = \exp\left(\sqrt{\sum_i \left(\ln(\mathrm{GSD}_{x_i}^2)\right)^2}\right) \quad (7)$$

In Eq. 7, we use the fact that the variance of each input variable is related to the corresponding $\mathrm{GSD}_{x_i}^2$ by $\mathrm{var}(\ln(x_i)) = \left(\ln(\mathrm{GSD}_{x_i}^2)\right)^2$. The choice of 2 in the exponent of the geometric standard deviations reflects the rounded critical value from the Student's *t*-distribution. All input variables are mutually independent – see Fantke et al. (2012b) for details. With that, relative sensitivities S_{x_i} are unity, i.e. $S_{x_i} = 1$, for all input variables and regression models (Slob 1994) and the uncertainty of model output exclusively depends on the variances of input variables and regression models. Considered in this analysis are pesticide-specific uncertainty factors for regression models and data for dissipation half-lives in crops (Fantke et al. 2014) representing the most uncertain variable in determining pesticide mass in crop harvest (Fantke et al. 2012b, Juraske et al. 2008), degradation half-lives in soil as proxy for soil residence times taken from the Pesticide Properties Database (Footprint 2014) or U.S. EPISuite (US-EPA 2012), crop-specific residue regression models for different harvest fraction ranges (Fantke et al. 2012b), post-harvest food processing (Keikotlhaile et al. 2010, Liang et al. 2014), fractions of applied pesticides lost to air and soil (DEFRA 2006), cancer and non-cancer dose-response information and severity factors (Huijbregts et al. 2005). $\mathrm{GSD}_{x_i}^2$ for all considered input variables and regression models are summarized in the SI, Section S-2. Since the harvest fraction regression model for each crop c in Eq. 6 involves an exponent of the complex form $\mathrm{hF}_{x,c,t} = 10^{(\alpha^*_{x,c} + \beta_{x,c} \times k_{x,c} \times t_x)}$, Eq. 7 was first applied within its domain of application to determine the 95% confidence interval of $\log(\mathrm{hF}_{x,c,t}) = \alpha^*_{x,c} + \beta_{x,c} \times k_{x,c} \times t_x$. The two-sided limits forming the confidence interval are then calculated as $\mathrm{hF}_{x,t} = \sum_c 10^{\log(\mathrm{hF}_{x,c,t})}$, yielding separate upper and lower 95% confidence interval limits at the level of harvest fractions, intake fractions and characterization factors.

3 Results

3.1 Intake fractions from pesticides applied to food crops

The variability of intake fractions for 875 pesticides applied to six crops is shown in <Figure **1**, contrasting the contributions of the fractions of applied pesticide reaching the agricultural food crops as residues and of the fractions reaching air and soil as emissions during and after application, of which the latter two are summed over all contributing exposure pathways. Intake fractions aggregated over crop residues and fractions lost to air and soil vary between 4 (tomato) and 6 (wheat, paddy rice, lettuce) orders of magnitude across pesticides applied to the same crop, demonstrating the importance of substance properties on crop residue dynamics. Aggregated intake fractions for the same pesticide applied to different crops vary between a factor 2.6 for herbicide florasulam and more than 5 orders of magnitude for 1-naphthol, a metabolite of insecticide carbaryl, demonstrating that the influences of crop characteristics and pesticide application times on crop residue dynamics are as important as the influence of substance properties. Individual intake fractions are provided for each of the 875 pesticides and six crops in the SI, Section S-3.

<Figure 1>

Highest aggregated intake fractions are found in lettuce and tomato with median values across pesticides of 0.035 and 0.013 $\mathrm{kg_{intake}\,kg_{applied}^{-1}}$, respectively, which is mostly due to very short averaged application times before harvest for insecticides and fungicides. In contrast, lowest aggregated intake fractions are found in potato with a median of 6×10^{-6} $\mathrm{kg_{intake}\,kg_{applied}^{-1}}$. The highest intake fractions for individual pesticide-crop combinations are found for fungicides cyproconazole and fuberidazole on lettuce yielding each 0.27 $\mathrm{kg_{intake}\,kg_{applied}^{-1}}$. Exposure from intake of crop residues is the main contributor to aggregated intake fractions for 88 to 97% of all pesticides in wheat, paddy rice, tomato, apple, and lettuce. For these crops, exposure from fractions lost to air and soil is the main contributor to aggregated intake fractions for only 1.3 to 7% of all pesticides (<Figure **1**, grey vs. white boxes). Potato is an exception, where exposure from fractions lost to air and soil is generally exceeding exposure from residues in treated crop and where the fraction lost to soil is the main contributor to aggregated intake fractions for 60% of the pesticides. For 35% of pesticides, the main contribution is from fractions lost to air and only 5% of all pesticides show main contribution from crop residues in potato.

The largest variability is shown for intake fractions associated with crop residues, which is mainly due to differences in application times between crops and between herbicides and other pesticides applied to the same crop. As an example, intake fractions associated with pesticides applied to apple trees show a variability of 6 orders of magnitude due to the large difference in average application times of 150 days (herbicides) and 14 days (non-herbicides) before crop harvest. Lower intake fraction ranges for all crops are associated with herbicides that are on average applied much longer before harvest than other pesticides, thereby allowing removal processes to limit crop residues at harvest time. In contrast, fungicides and insecticides are typically applied at later crop stages (sometimes even quickly before harvest) and therefore cover the upper range of crop residue-related intake fractions. Aggregated intake fractions for the individual pesticides and crops per kg applied are given in the SI, Section S-3, along with their 95% confidence intervals.

3.2 Human toxicological characterization factors for pesticides

Combining human intake fractions per mass of pesticide applied to different crops with toxicological effect information yields characterization factors shown in <Figure **2**. Whereas intake fractions could be derived for all 875 pesticides, cancer and non-cancer effect data are only available for a subset of 177 pesticides (20%) and 395 pesticides (45%), respectively. Hence, characterization factors are provided in the SI for a total of 465 pesticides associated with each of the six considered crops representing 53% of all pesticides included in this study.

<Figure 2>

Characterization factors for cancer-related effects typically show a lower variability than factors for non-cancer effects, which is linked to the fewer dose-response data available for cancer; hence, we provide carcinogenicity-related characterization factors only for a limited number of pesticides. In contrast, characterization factors for cancer effects seem to be more evenly distributed over the variability range than factors for non-cancer effects, most visible for lettuce and least visible for tomato (white boxes in <Figure **2**). This effect is linked to the influence of the variability of crop residue-related intake fractions (dominating overall human intake for all crops but potato) shown in <Figure **1**, where 50% of the data around the mean value for lettuce spread over more than 2 orders of magnitude, while 50% of data around the mean for tomato only differ by a factor 4.5. In line with this, the variability of characterization factors combining cancer and non-cancer effects (grey boxes in <Figure **2**) is influenced by the crop-specific variability of all contributing intake fractions (related to crop residues, air and soil fractions) and the variability of effect factors weighted by the number of contributing

data points (less for cancer, more for non-cancer effects). The highest variability of characterization factors is seen for lettuce with more than 9 orders of magnitude between lowest and highest factors of 4.4×10^{-9} $\mathrm{DALY\,kg_{applied}^{-1}}$ for ethanol and 9.3 $\mathrm{DALY\,kg_{applied}^{-1}}$ (attributable to the population-based cumulative risk of 3.4 non-cancer incidences $\mathrm{kg_{applied}^{-1}}$) for phenylmercuric acetate, respectively. Tomato shows the lowest variability in characterization factors of about 7 orders of magnitude ranging from 5.3×10^{-8} $\mathrm{DALY\,kg_{applied}^{-1}}$ for florasulam and 1.5 $\mathrm{DALY\,kg_{applied}^{-1}}$ (attributable to 1.6×10^{-3} cancer incidences and 3.9 non-cancer incidences $\mathrm{kg_{applied}^{-1}}$) for 2,4/2,6-toluene diisocyanate. Median values of characterization factors in aboveground crops across pesticides vary by less than a factor of 2 in descending order as lettuce > tomato > apple > paddy rice > wheat, whereas the median value for potato is 5 orders of magnitude lower. Characterization factors at midpoint level (cancer and non-cancer incidences $\mathrm{kg_{applied}^{-1}}$) and endpoint level (DALY $\mathrm{kg_{applied}^{-1}}$ given separately for cancer and non-cancer effects as well as aggregated over both) are given for all pesticide-crop combinations in the SI, Section S-3, including their 95% confidence intervals.

In Fig, we demonstrate along a realistic example how we arrived at characterization factors given in <Figure **2** and given per pesticide in SI (Section S-3), and how to apply human intake fractions and characterization factors in the context of LCA. We used as example pesticides tebuconazole and pyraclostrobin, both currently authorized e.g. in the European Union (European Commission 2011), and registered e.g. in Germany for use against leaf rust (*Puccinia recondita*) on wheat (BVL 2015).

<Figure 3>

3.3 Uncertainty in characterization modeling of pesticides

Uncertainty expressed as 95% confidence interval (CI) ranges is shown in Figure 4 for harvest fractions of 5250 pesticide-crop combinations. Confidence intervals are generally smallest for potato (median ratio of 97.5%-ile and 2.5%-ile CI limits of a factor 20), followed by tomato and apple (median 95% CI limit ratios of a factor 120 and 140, respectively), and are largest for lettuce and wheat (median 95% CI limit ratios of a factor 570 and 2680, respectively). The high end uncertainty for wheat is partly attributable to the long assumed time between application and crop harvest for herbicides (see SI, Section S-1). The much lower harvest fraction uncertainty ranges for pesticides applied to potato compared with applications to other crops is related to lacking uncertainty data for residence times in soil and, hence, does not indicate higher quality of regression models for potato.

Accuracy is in general higher in the range of high intake fractions and for the upper 97.5th percentile, whereas uncertainty grows with decreasing intake fractions as well as for the 2.5th percentile lower uncertainty limit. Accounting for improved estimates of half-lives is crops from Fantke et al. (2014) compared to earlier correlations from e.g. Juraske et al. (2008) has led to substantial changes and improvement in the accuracy of estimated harvest fractions (see SI, Figure S1). The range of pesticide half-lives in crops is now much narrower than the earlier estimates leading to a significant reduction in the variability between harvest fractions.

<Figure 4>

In all crops except potato, half-lives in/on crops along with time between pesticide application and crop harvest are the main contributor to crop residue dynamics. In case of potato the overall residence time in soil is the most influential factor that accounts for the various removal processes in the heterogeneous soil layer, before pesticides can enter the tuber via root uptake mechanisms (Juraske et al. 2011). Applying relatively large uncertainty to the crop-specific residues regression models compared with a relatively low generic uncertainty to soil degradation as proxy for overall soil residence time (most important for potato) yields generally lower uncertainty ranges of harvest fractions for potato than for all other crops. Harvest fractions for all pesticide-crop combinations are given in the SI, Section S-3, along with their 95% confidence intervals.

Despite uncertainty related to harvest fractions, uncertainty in subsequent human intake fractions (not shown) is scaled for each crop by the uncertainty associated with food processing factors, which were applied for each crop assuming a specific food processing step (see SI, Section S-1), but which were available as specific factors only for very few of the considered pesticides. Uncertainty from food processing, however, contributes on average only with 5.8% to intake fraction uncertainty across crops.

The highest share of characterization factor uncertainty with an average contribution of 70% is attributable to dose-response information, especially for extrapolated non-cancer effects. This is inherently limited by the availability of toxicity data for both risk assessment and LCA. Effective doses causing an effect in 50% of the exposed population have therefore mainly been extrapolated from toxicological studies with animals for which the specific health endpoints are mostly unknown, but derived from no-observed effect levels (NOEL). Using NOEL as starting point for estimating no-effect exposures leads to higher uncertainties (e.g. Landis & Chapman 2011) due to the large uncertainty around dose-response information. Since this type of effect information has been used in USEtox, we relied on the same data to ensure comparability across impact pathways and chemicals.

<Figure 5>

Figure 5 shows human health endpoint characterization factors for 465 pesticides with available effect information applied to 6 crops along with pesticide-specific uncertainty ranges that vary up to 9 orders of magnitude across pesticides applied to the same crop. Regarding the level of uncertainty and since uncertainty in the upper range of characterization factors is lower, Figure 5 is especially useful to provide an upper limit on the human health characterization factors and to identify with a food crop-related LCA study which pesticide(s) may provide a significant contribution compared to other life cycle impacts on human health associated with e.g. respiratory effects from exposure to fine particulate matter.

4 Discussion

4.1 Influences on intake fraction variability

Our results show that pesticide properties and crop characteristics are both strongly contributing to the variability of crop residues, fractions lost to air and soil, and subsequent human intake fractions of pesticides applied to agricultural food crops. We acknowledge that site characteristics, such as local soil and climate conditions during crop growth, and scenario characteristics, such as food processing and human consumption pattern might additionally contribute to the variability of our results, although to a lesser extent. The importance of crop characteristics, such as water content, growth, and leaf area index evolution, for crop residue dynamics is well in line with other studies demonstrating the strong influence of the choice of crop data on chemical distribution kinetics in crops (Trapp 2015) and on plant uptake dynamics from soil (Sun et al. 2014). Most importantly, the influence of all factors contributing to the variability of intake fractions from exposure to crop residues – the predominant component in the aggregated intake fractions from pesticide application to all considered crops except potato – is mostly associated with uncertainty of pesticide dissipation half-lives in crops. Uncertainty is additionally growing with increasing time between application and harvest. Accordingly, uncertainties around intake fraction values are also increasing with longer time to harvest allowing different uncertain model input variables to develop a significant influence on model output. However, with increasing time to harvest, intake fractions are typically lower, which makes the larger uncertainty less relevant than the (comparatively) lower uncertainty in the main range of interest, i.e. intake fractions $\geq 10^{-5}$ $\mathrm{kg_{intake}\,kg_{applied}^{-1}}$. This effect is shown in Figure 4 for harvest fractions as main driver of the magnitude of intake fractions. Uncertainty around fractions lost to air and soil along with associated intake fractions for emissions to air and soil are likely being underestimated in our study, since generic values for fractions lost and generic uncertainty for intake fractions have been used lacking more detailed data. This will not influence the general trends of our results, since these indirect contributions are low for most crops. Incorporating more realistic uncertainty values would nevertheless increase the variability of aggregated intake fractions for potato (with increasing time to harvest), where fractions lost to air and soil are dominating aggregated intake fractions for most pesticides (see <Figure **1**).

Overall, we reduced intake fraction variability between 1 and 9 orders of magnitude for pesticides applied to potato and apple, respectively, compared with estimates reported by Fantke et al. (2011b) for 121 pesticides (accounting for only 14% of the number of pesticides included in the present study). This reduction of variability in intake fractions is mainly attributable to improved dissipation data in crops. Overall, the uncertainty around intake fractions that is mainly driven by uncertainty in crop residues (Figure 4), is generally limited compared to uncertainty of characterization factors (Figure 5) that is strongly increased and dominated by uncertainty of (mainly non-cancer) dose-response information.

4.2 Accounting for realistic pesticide application

According to current national and international pesticide legislation we acknowledge that not all pesticides are allowed for use on all crops. In fact, there are many pesticides that are registered in some countries but banned for use in agriculture in other countries. Atrazine for example is a herbicide with endocrine disrupting properties (Hayes et al. 2011) that is one of the most widely used agricultural pesticides registered for use in the U.S. primarily on maize and sugarcane (US-EPA 2006), whereas its authorization in EU member states is withdrawn since 2004 (European Commission 2004). Given the heterogeneity in pesticide regulation between countries, we emphasize the need to verify the authorization status of all pesticides when applying our data. This is especially relevant when using our results for purposes of pesticide substitution and similar comparative assessments, where comparing two pesticides of which only one is registered for use on a specific crop could be misleading, if the unregistered pesticide shows lower intake fractions or characterization factors. Furthermore, we acknowledge that application times (days before crop harvest) are pesticide-crop combination-specific as a function of distribution dynamics in each crop-specific environment. In this study, we used application times before harvest that are averaged separately for herbicides and other pesticides to represent "typical" application times as best estimates for LCA that can also be applied to pesticides currently not included in our assessment. However, the uncertainty related to pesticide-specific application times before harvest for each crop (and country) is not included in our study and varies strongly between pesticides.

4.3 Data limitations and applicability in LCA studies

Our study shows several limitations. Experimental data for the most sensitive input variable, that is dissipation half-lives in crops, are only available for 311 out of 875 pesticides (35%). To account for the related uncertainty, we considered the higher uncertainty of the regression model to estimate crop dissipation to all pesticides, where experimental data were missing. The new correlations on half-lives have however substantially improved the accuracy of estimating related crop residues. Residence times in soil are the output of a generic system of mass balance equations accounting for the environmental fate of pesticides solved under the assumption of steady-state conditions with continuous emission input (Rosenbaum et al. 2008). Soil residence times are thereby influenced by a wide range of environmental characteristics including crop-related aspects and pesticide properties, of which degradation in soil plays an important role (Dubus et al. 2003). Lacking uncertainty data for soil residence times we applied uncertainty associated with soil degradation as proxy. We thereby acknowledge that we might underestimate the overall uncertainty specifically for potato, where soil residence time is driving the magnitude of crop residues and subsequent human intake. Whenever possible, soil degradation data are based on measurements aggregated in Footprint (2014) and only complemented by estimated data from the US-EPA (2012) when no experimental data were available. Differences in soil degradation data sources lead to differences in associated uncertainty, which were not considered in our study. We thereby acknowledge that data estimated from pesticide physicochemical properties may exceed measured field soil degradation half-lives by up to more than two orders of magnitude as can be seen when comparing e.g. tralomethrin or 8-quinolinol. These differences are becoming relevant in regulatory contexts, but are not as important in pure comparative assessments like LCA, where we are not bound to absolute thresholds for e.g. persistence in soil. Another limitation in our study is the use of generic fractions lost to air during and after pesticide application and associated uncertainty estimates. Further research is required to estimate these fractions more accurately in the context of LCA (Rosenbaum et al. 2015). However, for the majority of pesticide-crop combinations, this will not substantially influence related intake, since fractions lost to air are mostly not dominating intake fractions. Finally, we apply pesticide-specific data and averaged uncertainty factors for human health dose-response slope factors that are extrapolated from distinct exposed animal populations, exposure durations and routes and that are aggregated over a wide range of health endpoints (particularly for non-cancer effects). The difficulty to extrapolate effect factors from such inherently heterogeneous data leads to a significant contribution of dose-response information to overall uncertainty in characterization factors, which has already been acknowledged in previous studies (Huijbregts et al. 2005, Rosenbaum et al. 2008). While the quality of data underlying human toxicological effect factors needs to be improved accordingly, the variability of characterization factors across all pesticide-crop combinations spanning more than 9 orders of magnitude shows that relative to variability, overall uncertainty is not higher for toxicity-related impacts than for other impact categories.

Despite abovementioned limitations, our study contributes to significantly advancing the assessment of human health-related impacts from exposure to pesticides in LCA by including the predominant exposure pathway (i.e. intake of crop residues) and by improving the quality of the most uncertain input data for estimating pesticides in crop harvest (i.e. dissipation data in crops; see SI, Figure S1). Since our characterization factors are based on mass applied, LCA practitioners can and need to directly combine our results with pesticide application data as demonstrated in Fig. Whenever such data are not at hand, recommended application dosages as provided in The Pesticide Manual (Tomlin 2012) or on pesticide product labels can be applied as proxy.

5 Conclusions

We provide an operational framework for including human toxicity-related effects from exposure to pesticides via consumption of treated food crops into LCIA and provide for the first time uncertainty ranges around harvest fractions, intake fractions and characterization factors that are specific for each pesticide and crop. Results demonstrate that impacts of pesticides in terms of human toxicity are largely underestimated when ignoring exposure to residues in harvested and subsequently consumed crop components. For ready use in LCA studies, we present pesticide-crop combination-specific characterization factors normalized to pesticide mass applied and provide default data for application times and loss due to post-harvest food processing. Uncertainty needs to be considered when comparing results between different pesticides or with other chemicals to properly interpret ranking and maximum contributions, as it has been shown that pesticides with lower median characterization factors can be as important as pesticides with higher median characterization factors when considering the pesticide-specific uncertainty ranges. Improving dissipation half-lives in crops derived from experimental data has been essential in limiting uncertainties on harvest fractions. Further studies are required to better estimate fractions lost to air and soil during and after pesticide application and to reduce the inherent uncertainty in non-cancer toxicity effect information. When using our data, we emphasize the need to consult current pesticide regulation to allow for realistic scenarios where each pesticide is registered for use on certain crops only, which varies between countries.

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Figures

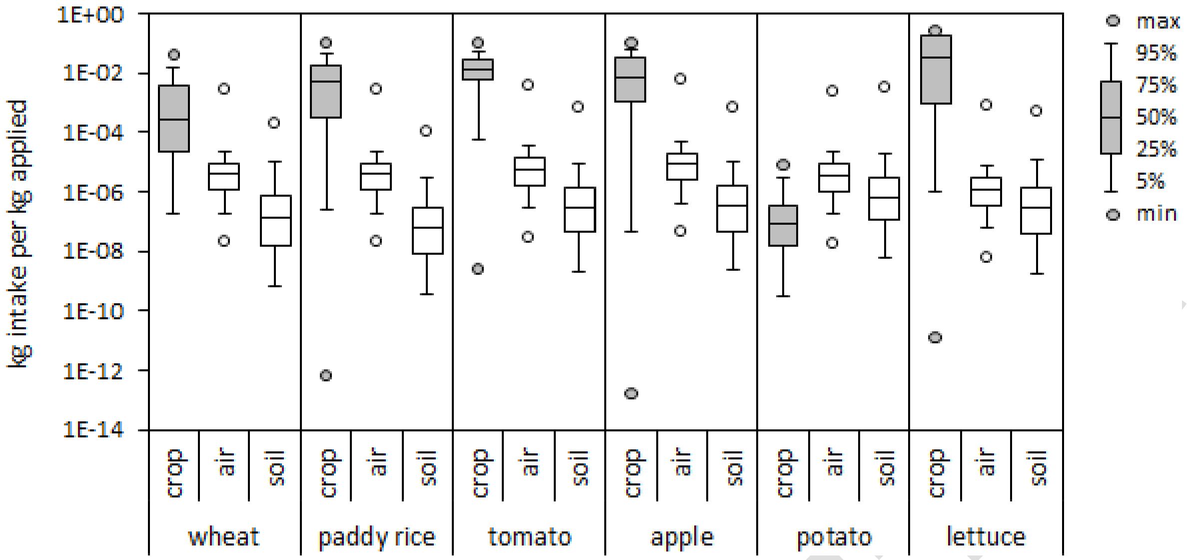

Figure 1 Variability of human intake fractions expressed as kg intake per kg applied pesticide grouped for each crop according to fractions reaching the crop as residues (grey boxes) and fractions reaching air and soil as emissions during and after application (white boxes). Minimum values below 10^{-14} are not displayed.

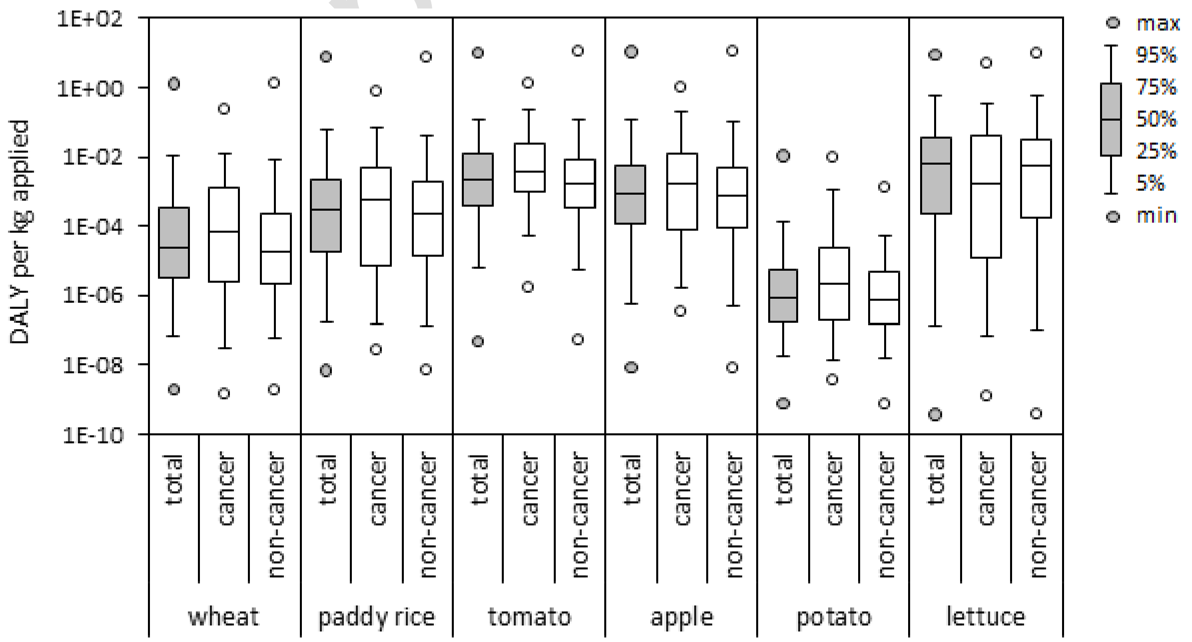

Figure 2 Variability of human toxicity endpoint characterization factors expressed as disability-adjusted life years (DALY) per kg applied pesticide grouped for each crop into total aggregated effects (grey boxes), and cancer and non-cancer effects (white boxes).

(a) human intake = mass applied per functional unit × total intake fraction

$$\text{human intake (tebuconazole)} = 0.25\ \text{kg}_{\text{applied}}/\text{ha} \times 7.1\times10^{-4}\ \text{kg}_{\text{intake}}/\text{kg}_{\text{applied}} = 1.8\times10^{-4}\ \text{kg}_{\text{intake}}/\text{ha}$$

$$\text{human intake (pyraclostrobin)} = 0.125\ \text{kg}_{\text{applied}}/\text{ha} \times 2.0\times10^{-5}\ \text{kg}_{\text{intake}}/\text{kg}_{\text{applied}} = 2.2\times10^{-6}\ \text{kg}_{\text{intake}}/\text{ha}$$

(b) characterization factor* = total ingestion intake fraction × non-cancer dose-response for ingestion × non-cancer severity factor

$$\text{CF (tebuconazole)} = 7.1\times10^{-4}\ \text{kg}_{\text{ingested}}/\text{kg}_{\text{applied}} \times 0.015\ \text{incidences}_{\text{non-cancer}}/\text{kg}_{\text{ingested}} \times 2.7\ \text{DALY}/\text{incidence}_{\text{non-cancer}} = 2.9\times10^{-5}\ \text{DALY}/\text{kg}_{\text{applied}}$$

$$\text{CF (pyraclostrobin)} = 2.0\times10^{-5}\ \text{kg}_{\text{ingested}}/\text{kg}_{\text{applied}} \times 0.042\ \text{incidences}_{\text{non-cancer}}/\text{kg}_{\text{ingested}} \times 2.7\ \text{DALY}/\text{incidence}_{\text{non-cancer}} = 2.2\times10^{-6}\ \text{DALY}/\text{kg}_{\text{applied}}$$

(c) health impact = mass applied per functional unit × characterization factor

$$\text{health impact (tebuconazole)} = 0.25\ \text{kg}_{\text{applied}}/\text{ha} \times 2.9\times10^{-5}\ \text{DALY}/\text{kg}_{\text{applied}} = 7.3\times10^{-6}\ \text{DALY}/\text{ha}$$

$$\text{health impact (pyraclostrobin)} = 0.125\ \text{kg}_{\text{applied}}/\text{ha} \times 2.2\times10^{-6}\ \text{DALY}/\text{kg}_{\text{applied}} = 2.8\times10^{-7}\ \text{DALY}/\text{ha}$$

*In this example, we calculated the characterization factors exclusively from ingestion intake fractions (inhalation intake fractions contribute to overall intake fraction only with 0.02% for tebuconazole and 0.32% for pyraclostrobin) and non-cancer dose-response (cancer effect data were not available). Whenever inhalation intake fractions and/or cancer effects become relevant, they need to be included in the characterization factor calculations.

Figure 3 Calculation steps for deriving human intake per treated hectare (a), endpoint characterization factors (b), and health impacts per treated hectare (c) for two example fungicides applied to wheat. Tebuconazole is typically applied as 250 g/l emulsion at 1 l/ha (Bayer 2014) and pyraclostrobin is typically applied as 250 g/l emulsion at 0.5 l/ha (BASF 2012). Intake fractions, dose-response factors and characterization factors are given in SI (Section S-3).

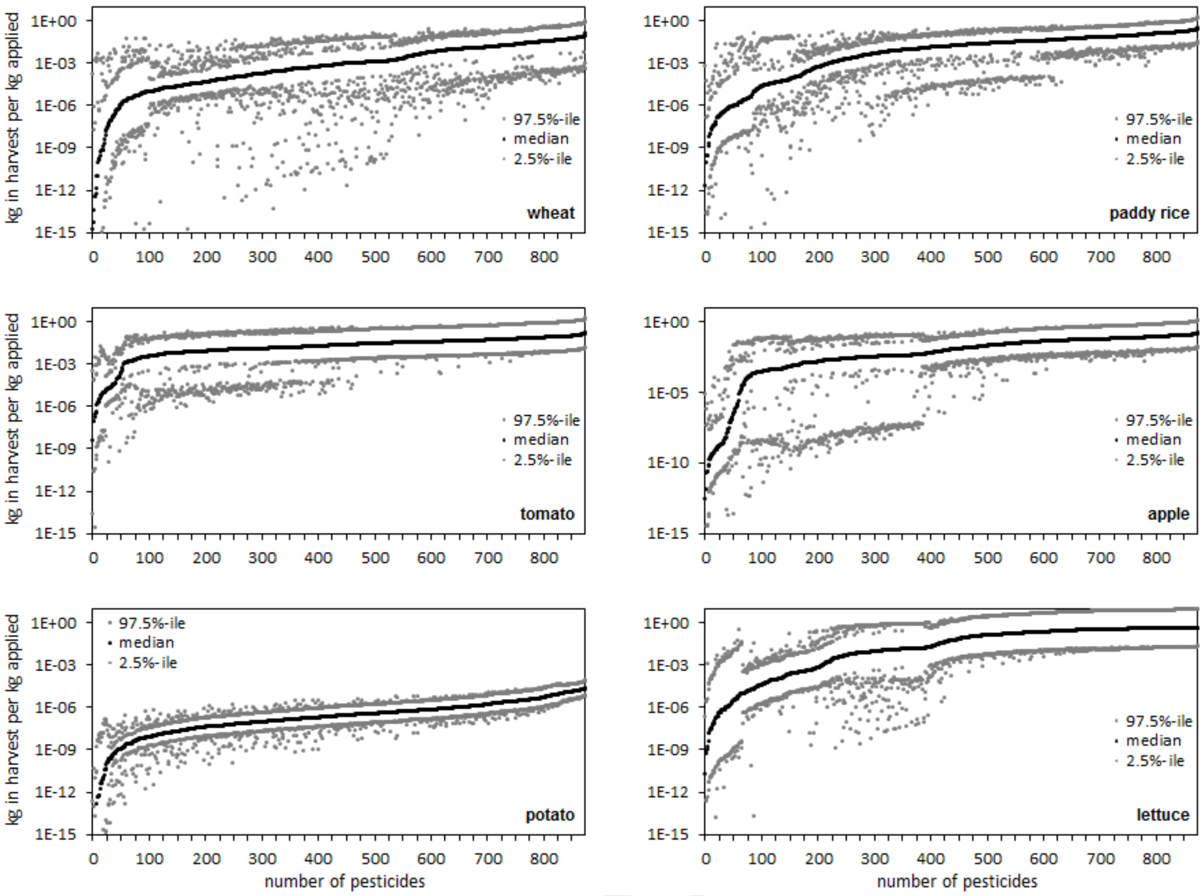

Figure 4 Uncertainty of harvest fractions for 875 pesticides and 6 crops expressed as 95% confidence interval ranges of pesticide mass in crop harvest per kg applied pesticide.

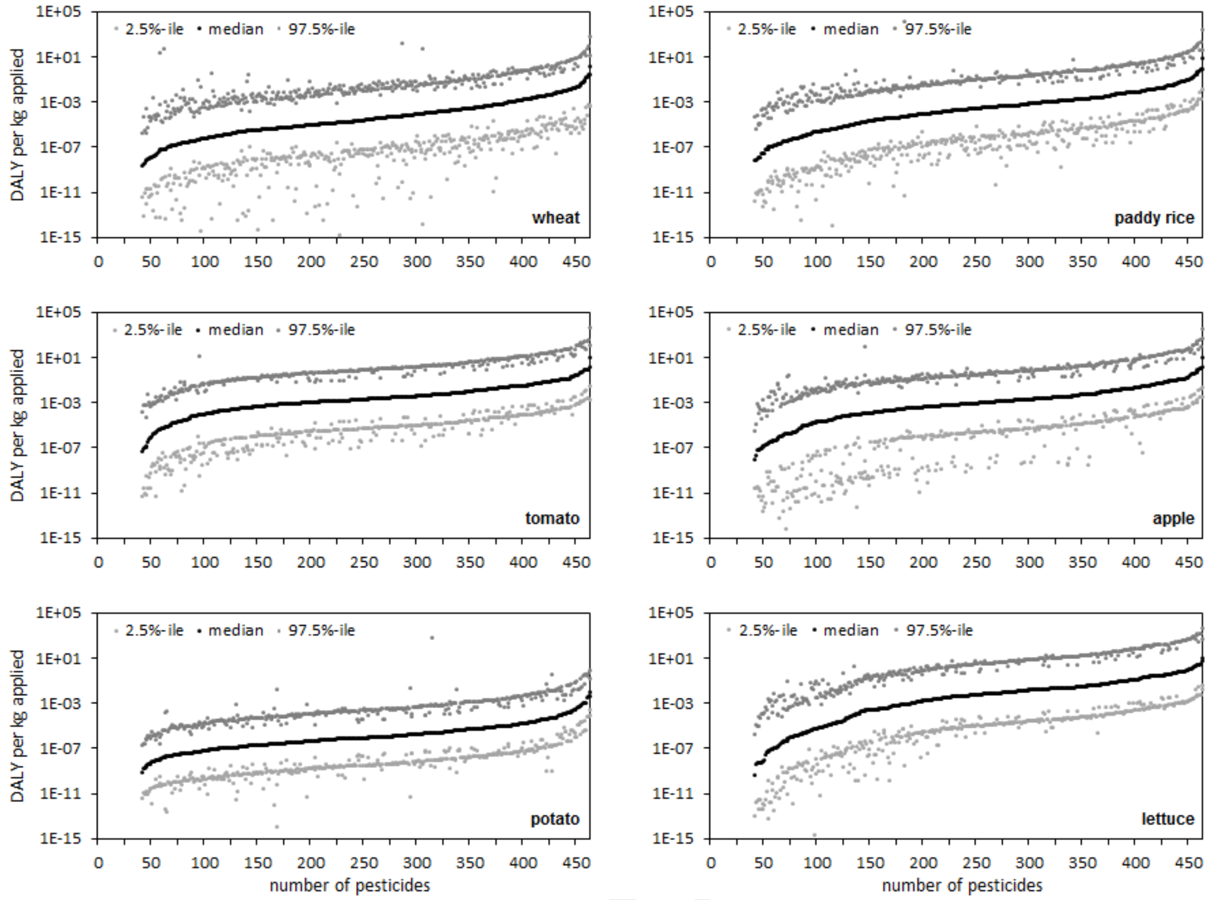

Figure 5 Uncertainty of human toxicological characterization factors at endpoint level for the reduced set of 465 pesticides with available toxicity effect information and 6 crops expressed as 95% confidence interval ranges of disability-adjusted life years (DALY) per kg applied pesticide.